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**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES**

Inventor/Appellant: Partho Sarkar

Title: CRACK-RESISTANT ANODE-SUPPORTED FUEL CELL

Application No. 10/658,803

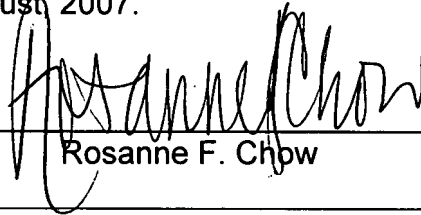
Filed: September 9, 2003

Examiner/Art Unit: ALEJANDRO, Raymond / 1745

Attorney File No.: 2281-001-03

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Rosanne F. Chow

RESPONSE TO NOTIFICATION OF NON-COMPLIANT APPEAL BRIEF

Dear Commissioner for Patents:

In response to a Notification of Non-Compliant Appeal Brief mailed July 31, 2007 in the above-referenced patent application, an amended appeal brief is hereby submitted. Changes to the Appeal Brief filed on February 8, 2007 that are contained in the amended appeal brief are shown in the enclosed redlined version.

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Please contact the undersigned at 425-455-5575 if with any questions or comments regarding this matter. If the Patent Office finds that any fees are due, please charge any additional fees or credit overpayment to Deposit Account No. 07-1897.

Dated: August 31, 2007

Respectfully submitted,



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Enclosures:

Copy of Notification of Non-Compliant Appeal Brief dated July 31, 2007
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Rosanne F. Chow

APPEAL BRIEF

Dear Commissioner for Patents:

This is an appeal pursuant to 37 C.F.R. § 1.191(a) from the decision of the Examiner, dated June 5, 2006, finally rejecting claims 1-12 of the above-referenced patent application. A request for a one-month extension of time is enclosed pursuant to 37 C.F.R. 1.36(a).

Attached to this Brief as **Appendix A** is a claims appendix containing a copy of all the claims involved in the Appeal, as required under 37 C.F.R. §41.37(a)(viii). An evidence appendix as required under 37 C.F.R. §41.37(a)(ix) accompanies this Brief in the form of **Appendices B-E**. **Appendix B** contains pages from a publication that illustrates the oxygen ion conduction and high electrical conductivity characteristics of the solid electrolyte yttria stabilized zirconia ("YSZ") that make YSZ suitable for use in

the anode support layer of a solid oxide fuel cell ("SOFC"). **Appendix C** contains pages from a publication illustrating the relatively high electrical resistivity of alumina (aluminum oxide). **Appendix D** contains pages from a publication further discussing desirable characteristics of solid electrolytes suitable for use in SOFCs. **Appendix E** lists several Web sites that contain exemplary information illustrating the characterization and use of alumina as an insulator.

The objective evidence that accompanies this brief as **Appendices B-E** was made of record in the Response to final Office Action filed on October 11, 2006. This evidence was submitted after the final Office Action because this was when the Examiner first requested such evidence (See page of the Interview Summary mailed September 12, 2006). Thus, this evidence was necessary to fully respond to the Examiner's request and could not have been earlier presented since such a request had not been made until that point in the prosecution of the present application. These are certainly good and sufficient reasons pursuant to 37 C.F.R. § 1.116(e) and this evidence is therefore of record in the present application in accordance with 37 C.F.R. §41.37(c)(2).

A related proceedings appendix as required under 37 C.F.R. §41.37(c)(1)(ii) accompanies this brief as **Appendix F** and indicates there are no related appeals, interferences, or judicial proceedings, as is discussed in more detail in Section II below.

I. *Real Party In Interest*

The real party in interest is Alberta Research Council, Inc., having a principal place of business at 250 Karl Clark Road, Edmonton, Alberta T6N 1E4, Canada. The inventors assigned this application to Alberta Research Council, Inc. ("Assignee") in an Assignment recorded on February 9, 2004 at reel no. 014958, frame no. 0347.

II. *Related Appeals and Interferences*

Based on information obtained from Alberta Research Council, Inc., and based on information and belief of the undersigned attorney, there are no prior or pending

appeals, interferences, or judicial proceedings known to Appellant, Appellant's legal representative the undersigned attorney, or the Assignee, which are related to, which directly affect or which will be directly affected by, or which have a bearing on the decision of the Board of Patent Appeals and Interferences ("the Board") in the pending Appeal. There are no such appeals, interferences, or judicial proceedings as indicated in the related proceedings appendix that accompanies this brief as **Appendix F**.

III. Status of Claims

Claims 1-13 are pending and stand rejected by the Examiner. Claims 1-12 are appealed.

IV. Status of Amendments

An Amendment After Final was filed on October 11, 2006 in response to a Final Office Action mailed on June 5, 2006. In an Advisory Action mailed on October 17, 2006, the Examiner indicated the proposed amendments in the Amendment After Final would not be entered as raising new issues that would require further consideration and/or search. **Appendix A** includes all the appealed claims 1-12 as they are currently pending.

V. Summary of Claimed Subject Matter

This summary of the invention provides cross-referencing to the application as required by 37 C.F.R. § 41.37(c)(v). This cross-referencing is solely to assist the Board in understanding the Applicant's invention and is not meant to be exhaustive or to limit the scope of the pending claims.

Independent claim 1 recites an anode-supported solid oxide fuel cell including an anode support layer comprising a porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material. An electrolyte layer is in adjacent intimate contact with the anode support layer and a cathode layer is in adjacent intimate contact with the electrolyte layer.

Figure 1 illustrates a planar anode-supported solid oxide fuel cell 2 corresponding to one embodiment of present invention covered by claim 1. The anode-supported solid oxide fuel cell recited in claim 1 corresponds to the fuel cell 2 in the embodiment of Figure 2. See page 5, lines 25-30 and page 6, lines 1-3 (paragraph 16). The anode support layer recited in claim 1 corresponds to an anode support layer (ASL) 16 in the embodiment of the fuel cell 2 shown in Figure 2. *Id.* Claim 1 further recites that the anode support layer includes a porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material. This recited porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material is discussed on page 6, lines 16-30 and page 7, lines 1-2 (paragraph 19) with reference to the ASL 16. Claim 1 further recites an electrolyte layer that is in adjacent intimate contact with the anode support layer, where the recited electrolyte layer corresponds to an electrolyte layer 12 in the embodiment of Figure 1. *Id.* Finally, claim 1 recites a cathode layer that is in adjacent intimate contact with the electrolyte layer, where the recited cathode layer corresponds to a cathode layer 10 in the embodiment of Figure 2. *Id.*

Turning now to independent claim 12, this claim recites an anode-supported solid oxide fuel cell including a cathode layer in adjacent intimate contact with an electrolyte layer that is in adjacent intimate contact with an anode functional layer. The anode functional layer is in adjacent intimate contact with an anode support layer comprising an ion-conducting structure with a plurality of vias extending through the thickness of the oxygen ion-conducting structure, with at least some of the vias being filled with electronically conductive material.

Claim 12 covers, for example, the embodiment of the present invention illustrated in Figure 2. The anode-supported solid oxide fuel cell recited in claim 12 corresponds to the fuel cell 2 in the embodiment of Figure 2. See page 8, lines 22-30 and page 9, lines 1-3 (paragraph 24). Claim 12 further recites that the fuel cell includes a cathode layer in adjacent intimate contact with an electrolyte layer that is in adjacent intimate contact with an anode functional layer. The recited cathode layer, electrolyte layer, and anode functional layer correspond to a cathode layer 10, electrolyte layer 12,

and anode functional layer 14, respectively, in the embodiment of Figure 2. Claim 12 further recites that the anode functional layer, which corresponds to layer 14 in Figure 2, is in adjacent intimate contact with an anode support layer comprising an ion-conducting structure with a plurality of vias extending through the thickness of the oxygen ion-conducting structure, with at least some of the vias being filled with electronically conductive material. The recited anode support layer corresponds to the anode support layer 16 in the embodiment of Figure 2. *Id.* The recited plurality of vias extending through the thickness of the oxygen ion-conducting structure correspond to the vias 20 in the embodiment of Figure 2. *Id.*

VI. *Grounds of Rejection to Be Reviewed On Appeal*

- 1. Whether U.S. Patent Application Publication No. 2002/0164523 to Shibata et al. (“Shibata”) anticipates claims 1-6, 9, and 11-12 under 35 U.S.C. § 102(e).**
- 2. Whether claims 7-8 and 10 are obvious under 35 U.S.C. § 103(a) in view of the combination of Shibata and U.S. Patent Application Publication No. 2002/0028367 to Sammes et al. (“Sammes”).**

VII. *Argument*

- 1. Claims 1-6, 9, and 11-12 are allowable under 35 U.S.C. § 102(e) over U.S. Patent Application Publication No. 2002/0164523 to Shibata et al. (“Shibata”) because Shibata neither discloses nor suggests an anode support layer comprising a porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material.**

“Under 35 U.S.C. § 102, every limitation of a claim must identically appear in a single prior art reference for it to anticipate the claim.” *Gechter v. Davidson*, 42

USPQ2d 1030, 1032 (Fed. Cir. 1997). Before determining whether a prior art reference anticipates a claim, one must first construe the claim “to define the scope and meaning of each contested limitation.” *Id.* The proper starting place in any claim construction analysis is the claim language itself read in view of the specification and the prosecution history. *Phillips v. AWH Corp.*, 415 F.3d 1303 (Fed. Cir. 2005) (*en banc*).

In the final Office Action mailed 5 June 2006, the Examiner maintained the rejections of claims 1-6, 9, and 11-12 under 35 U.S.C. § 102(e) as being anticipated by Shibata. On September 7, 2006, the undersigned along with the inventor Partho Sarkar and Brian Y. Lee, Canadian counsel for the Assignee, held a telephone interview with the Examiner to discuss the rejections of the pending claims (See the Examiner's Interview Summary mailed September 12, 2006 that is part of the file history of the present application). During this telephone interview, the Examiner construed the “porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material” recited in claim 1 as corresponding to the porous metallic base body 1 illustrated in Figures 4-7. Paragraph 44 of Shibata states “it may be possible for the porous metallic base body [1] to be made of a ceramic which is plated with the above metals [e.g., nickel Ni] or with the alloy containing the above metals.” Paragraph 69 more specifically provides “the porous base body 1 ... includes a ceramic (alumina) body plated with [nickel] Ni.” In both the final Office Action (Item 2, bottom of page 3) and during the telephone interview, the Examiner erroneously concluded that the alumina portion of the base body 1 corresponds to the “porous ion-conducting structure” and the nickel Ni coating corresponds to the “catalytic and electronically conductive material” as recited in claim 1.

Notwithstanding the Examiner's assertions, alumina is not a porous ion-conducting material. This fact is well understood and known by those skilled in the art. Alumina is an insulator having a resistivity that is too high for this material to be considered a solid electrolyte and an oxygen ion conductor suitable for use in a solid oxide fuel cell. In order for a material to be considered an oxygen ion conductor suitable for use in an electrode of a solid oxide fuel cell, the material must provide the necessary and efficient ionic path for anodic reaction which takes place during solid

oxide fuel cell operation. As is well known in the solid oxide fuel cell art, the ionic conductivity of the anode must be comparable to the ionic conductivity of the electrolyte. Moreover, alumina's resistivity is too high at the operating temperature of a solid oxide fuel cell to perform the required electrode function for the fuel cell.

During the Examiner interview, the Examiner contended that even if alumina is considered in the prior art as an insulating material this material is nonetheless capable of conducting some ions, even if such ion conductivity is poor. See page 3 of the Interview Summary. The Examiner requested objective evidence to support that such an alumina ceramic is "incapable" of transporting ions, meaning that the material exhibits no ion conduction at all. *Id.*

A suitable material for the recited anode support layer and thus an ion-conducting material is yttria stabilized zirconia (YSZ). See, e.g., paragraph 8 of the present application. YSZ is an excellent conductor of negatively charged oxygen (oxide) ions at high temperatures. See paragraph 3. A solid oxide fuel cell operates at an elevated temperature, typically on the order of between 700-1000 °C. *Id.* See page 101 of the Suresh publication that accompanies this amendment as **Appendix B** regarding general characteristics of ion-conducting materials. Other oxygen ion conducting materials suitable for use in a solid oxide fuel cell may be substituted for YSZ in the anode support layer. See paragraph 19. A characteristic of a solid electrolyte, which may otherwise be known as a fast ion conductor or a superionic solid, is a high electrical conductivity in the range of 10^{-1} - 10^{-4} ohm⁻¹cm⁻¹ (*i.e.* a resistivity of 10-10,000 ohm⁻¹cm⁻¹). See page 17 of **Appendix B**. In contrast, alumina (aluminum oxide) has a resistivity of 5.0×10^8 at 700°C and 2×10^6 at 1000°C (see page 959 of the Shackelford and Alexander publication, which accompanies this amendment as **Appendix C**. This large resistivity of alumina plainly evidences that alumina is not a solid electrolyte/fast ion conductor/superionic solid suitable for use in a solid oxide fuel cell. See the highlighted portions of the article that accompanies this amendment as **Appendix D** for additional information regarding desired resistivity values in fuel cells. **Appendix E** contains a listing of several Web sites that illustrate alumina being characterized and used as an insulator and not an ionic-conductor.

All the above evidence clearly demonstrates that the Examiner is in error in asserting that the alumina disclosed in Shibata corresponds to the recited porous ion-conducting material. Moreover, the Examiner's contention that because alumina must exhibit some ion conductivity the base body 1 of Shibata may be considered to correspond to the porous ion-conducting structure of the anode support layer recited in claim 1. Materials are classified as having physical characteristics that result in each material being placed in a particular class of materials, such as an electric or thermal conductor or insulator or an ionic conductor or insulator. Materials are not classified in absolute terms as would be required using the Examiner's logic. The Examiner requested evidence that "ceramic alumina is INCAPABLE of transporting ions (no ion conduction at all)." See page 3 of the Interview Summary. No such evidence can be provided for any material. Although classified as a particular type of material, every material will exhibit some characteristics of another class of materials. For example, materials classified as electrical insulators exhibit some amount of electrical conductivity, but such conductivity is so small that these materials are nonetheless classified as insulators. If the Examiner's argument was accepted, then the classification of materials would be rendered meaningless. Any material could be said to be whatever type of material was needed by an Examiner when formulating a rejection.

Pursuant to MPEP § 2111, during patent examination the pending claims must be given their broadest reasonable interpretation consistent with the specification. The Examiner expressly mentioned this well known tenet of patent examination during the Examiner interview. As expressly set forth in Section 2111, the "broadest reasonable interpretation of the claims must also be consistent with the interpretation that those skilled in the art would reach." As evidenced by the accompanying technical literature, one skilled in the art would not interpret the phrase "ion-conducting structure" to include the insulating material of alumina ceramic. The Examiner's attempt to so interpret this phrase is simply put an unreasonable interpretation of this language. Although the Examiner cites the "broadest reasonable interpretation" language of Section 2111 as the rationale for his interpretation, such an interpretation of this language plainly in violation of this section of the MPEP as well as in contravention of common sense.

For all these reasons, Shibata neither discloses nor suggests an anode support layer comprising a porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material as recited in claim 1. The base body 1 of Shibata simply cannot be said to correspond to the recited anode support layer. As discussed above, an alumina ceramic simply is not “a porous ion-conducting structure.”

Because an alumina ceramic exhibits some ion conductivity does not make such an alumina ceramic an ion-conducting structure. As previously discussed with reference to **Appendices B-E**, alumina’s resistivity is too high for this material to be considered a solid electrolyte and an ion conductor for a solid oxide fuel cell. Alumina’s resistivity is also too high at the operating temperatures of solid oxide fuel cells to carry out the required electrode function.

Furthermore, claim 1 recites an electrolyte layer in intimate contact with the anode support layer. Shibata discloses electrodes 10 and 11 to be in intimate contact with the electrolyte and not the base body 1 asserted by the Examiner to correspond to the anode support layer. Thus, claim 1 is allowable for this additional reason.

For all these reasons, the combination of elements recited in claim 1 is allowable and dependent claims 2-11 are allowable for at least the same reasons as claim 1 and due to the additional limitations added by each of these claims.

Claim 12 recites an anode-supported solid oxide fuel cell comprising an anode support layer including an ion-conducting structure with a plurality of vias extending through the thickness of the oxygen ion-conducting structure. At least some of the vias are filled with electronically conductive material. An anode functional layer is in adjacent intimate contact with the anode support layer and an electrolyte layer is in adjacent intimate contact with the anode functional layer. A cathode layer is in adjacent intimate contact with the electrolyte layer.

Shibata neither discloses nor suggests an anode support layer as recited in claim 12. The base body 1 of Shibata simply cannot be said to correspond to the recited anode support layer since an alumina ceramic is not an oxygen ion-conducting structure. Simply because an alumina ceramic exhibits some ion conductivity does not make such an alumina ceramic an ion-conducting structure. Alumina’s resistivity is too

high for this material to be considered a solid electrolyte and an oxygen ion conductor for a solid oxide fuel cell and the resistivity is also too high at the operating temperatures to carry out the required electrode function.

For all these reasons, the combination of elements recited in claim 12 is allowable.

During the interview, the Examiner also raised what he termed a potential 35 U.S.C. § 112, paragraph 1, issue regarding the recitation of an ion-conducting structure in the claims and pointed to paragraph 19 of the specification to support his assertion that only a description of oxygen ion conducting materials is contained in the application. See page 3 of the Interview Summary. The Examiner states that “the invention, as presently claimed, may have a [Section] 112 issue (lack of adequate written support, and may be [sic] lack of enablement) because it clearly does not disclose, suggest or teach how “ANY” ion conducting material can be suitably used in the claimed anode structure.” While not at issue here a brief reply to this comment is provided. It is the Examiner and not the Applicant that is inserting the word “any” before the claim term “ion-conducting structure” recited in claims 1 and 12. Claim terms must be construed in the context of the application. As expressly noted by the Examiner on page 3 of the Interview Summary, the present application (paragraph 19, lines 4-7) states “other oxygen ion conducting materials suitable for SOFC use and having a relatively similar thermal coefficient to the electrolyte, as is known in the art, may be substituted for YSZ.” Claims 1 and 12 are directed to anode-supported solid oxide fuel cells. In the context of solid oxide fuel cells, one skilled in the art would understand the recited term “ion conducting structure” to relate to oxygen ions. No Section 112 issue exists.

- 2. Claims 7-8 and 10 are nonobvious under 35 U.S.C. § 103(a) in view of the combination of Shibata and U.S. Patent Application Publication No. 2002/0028367 to Sammes et al. (“Sammes”) since there is no disclosure or suggestion.**

In the final Office Action, the Examiner rejected claims 7, 8 and 10 under 35 U.S.C. § 103(a) as being obvious in view of Shibata and Sammes. As discussed

above, nothing in Shibata discloses or suggests the recited anode support layer. Furthermore, Sammes describes anode layers that comprise different ratios of electrochemically active substance. Sammes also describes a process for manufacturing a solid oxide fuel cell anode wherein YSZ powder is added to NiO powder and these materials are milled, extruded, dried and sintered together. As discussed above, one would expect a cross-sectional micrograph of the base body described in Sammes to reveal a porous structure comprising a homogenous composition within each layer. There is no suggestion in Sammes to impregnate catalytic and electronically conductive material into the pores of an anode support layer, thereby creating an anode support layer having a non-homogeneous porous structure.

Claim 7 recites the fuel cell of claim 5 wherein the catalytic and electronically conductive material is Ni-containing material and is compositionally graded through the thickness of the anode support layer, with a higher concentration of the Ni-containing material at one major surface of the anode support layer than the other. The recited porous structure is non-homogeneous in that the electronically conductive material of nickel Ni is compositionally graded through the thickness of the anode support layer. As a result, the anode support layer has a higher concentration of nickel Ni at one major surface than at the other major surface. Within the recited single anode support layer the electronically graded material is graded through the thickness of this single layer. Sammes discloses multiple layers with the concentration within each layer being homogeneous or constant and not graded. For these reasons, even if combined Shibata and Sammes do not teach or suggest the recited structure of claim 7. Claim 7 is accordingly allowable.

Claim 8 recites the fuel cell of claim 7 wherein the anode support layer further comprises a second conductive metal selected from the group of ferritic steel, super alloy, and Ni-Ag alloy and which is concentrated at the major surface of the anode support layer having the lower concentration of Ni-containing material. Once again, Sammes neither discloses or suggest a single anode support layer having a nickel Ni concentration graded as recited in claim 7 and then a second conductive material concentrated at the major surface of the anode support layer having the lower

concentration of nickel Ni as recited in claim 8. Claim 8 is accordingly allowable for these additional reasons.

3. Dependent Claims Not Specifically Addressed In Section VIII

All dependent claims not specifically addressed in this section are patentable by virtue of their respective dependencies from claims for which the Applicants have presented an argument for patentability.

VIII. *Claims Appendix*

Appendix A includes all the appealed claims 1-12 as they are currently pending.

IX. *Evidence Appendix*

Appendices B through E include all objective evidence that was made of record in the Response to Final Office Action filed on October 11, 2006.

X. *Related Proceedings Appendix*

Appendix F indicates there are no related appeals, interferences, or judicial proceedings, as is discussed in more detail in Section II above.

XI. *Conclusion*

For the foregoing reasons, the Applicants request the Board to reverse the Examiner's rejection of claims 1-6, 9, and 11-12 under 35 U.S.C. § 102(e) and his rejection of claims 7, 8, and 10 under 35 U.S.C. § 103(a) and remand the application to the Examiner for issuance of claims 1-12.

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Dated: August 31, 2007

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Enclosures:

Appendices A-F
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APPENDIX A

1. An anode-supported solid oxide fuel cell comprising
 - (a) an anode support layer comprising a porous ion-conducting structure having pores impregnated with a catalytic and electronically conductive material;
 - (b) an electrolyte layer in adjacent intimate contact with the anode support layer; and
 - (c) a cathode layer in adjacent intimate contact with the electrolyte layer.
2. The fuel cell of claim 1 wherein the catalytic and electronically conductive material is selected from the group of nickel, copper, silver, tungsten, and any alloys of these materials.
3. The fuel cell of claim 2 further comprising a second phase material mixed with the catalytic and electronically conductive material, the second phase material being selected from the group of yttria-stabilized zirconia (YSZ), doped cerium oxide, alumina or its salts.
4. The fuel cell of claim 2 further comprising an anode functional layer between the anode support layer and the electrolyte layer such that the electrolyte layer is in adjacent intimate contact with the anode functional layer instead of the anode support layer.
5. The fuel cell of claim 4 wherein the porous ion-conducting structure of the anode support layer is substantially yttria-stabilized zirconia (YSZ).

6. The fuel cell of claim 5 wherein the catalytic and electronically conductive material is substantially evenly distributed throughout the anode support layer.
7. The fuel cell of claim 5 wherein the catalytic and electronically conductive material is Ni-containing material and is compositionally graded through the thickness of the anode support layer, with a higher concentration of the Ni-containing material at one major surface of the anode support layer than the other.
8. The fuel cell of claim 7 wherein the anode support layer further comprises a second conductive metal selected from the group of ferritic steel, super alloy, and Ni-Ag alloy and which is concentrated at the major surface of the anode support layer having the lower concentration of Ni-containing material.
9. The fuel cell of claim 4 further comprising a porous zirconia-nickel cermet buffer layer sandwiched in between the anode support layer and anode functional layer, and having a porosity between 40-90%.
10. The fuel cell of claim 4 wherein the porous ion conducting structure of the anode support layer is comprised of a mixture of 10-30 vol. % of Ni, or NiO or both, and the balance yttria-stabilized zirconia (YSZ).
11. The fuel cell of claim 4 wherein the anode support layer further comprises a plurality of vias extending through the thickness of the ion conducting structure of the anode support layer, at least some of the vias being filled with an electronically conducting material.
12. An anode-supported solid oxide fuel cell comprising

- (a) an anode support layer comprising an ion-conducting structure with a plurality of vias extending through the thickness of the oxygen ion-conducting structure, at least some of the vias being filled with electronically conductive material;
- (b) an anode functional layer in adjacent intimate contact with the anode support layer;
- (c) an electrolyte layer in adjacent intimate contact with the anode functional layer; and
- (d) a cathode layer in adjacent intimate contact with the electrolyte layer.

13. An anode-supported solid oxide fuel cell comprising
- (a) an anode support layer;
 - (b) a porous cermet buffer layer in adjacent intimate contact with the anode support layer, and being composed of a zirconia-nickel cermet with a porosity between 40 and 90%;
 - (c) an anode functional layer in adjacent intimate contact with the buffer layer;
 - (d) an electrolyte layer in adjacent intimate contact with the anode functional layer; and
 - (e) a cathode layer in adjacent intimate contact with the electrolyte layer.

APPENDIX B

*super-
ionic
solids*

PRINCIPLES
AND
APPLICATIONS

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CHAPTER 2

Superionic Materials and Structures

Superionic solids are ionic materials with high electrical conductivity comparable with those of liquid electrolytes. These materials are also termed "solid electrolytes" or "fast ion conductors". Typically a superionic solid has the following characteristics:

- (i) crystal bonding is ionic;
- (ii) electrical conductivity is high ($10^{-1} - 10^{-4} \text{ ohm}^{-1} \text{ cm}^{-1}$);
- (iii) principal charge carriers are ions which means that the ionic transference number (t_{ion}) is almost equal to 1 (here t_{ion} refers to the fractional contribution of the ionic conductivity to the total conductivity);
- (iv) the electronic conductivity is small; generally materials with electronic transference number (t_e) less than 10^{-4} are considered satisfactory superionic solids.

The values of electrical conductivity of a few ionic and superionic solids are shown in Figure 2.1. The highest conductivity at room temperature obtained so far is for RbAg_4I_5 which is $0.27 \text{ ohm}^{-1} \text{ cm}^{-1}$. This is many orders of magnitude higher than those of the more commonly known ionic solids KCl , NaCl etc, which have room temperature conductivity $\sim 10^{-12} - 10^{-16} \text{ ohm}^{-1} \text{ cm}^{-1}$. It may be noted that most of the superionic materials attain a high electrical conductivity above a certain temperature which may or may not be well defined. In other words, with increasing temperature the electrical conductivity sometimes changes gradually (as in β -alumina) or shows an abrupt jump (e.g. in β - AgI , RbAg_4I_5 etc.). Furthermore, for the latter materials the abrupt conductivity change is sometimes associated with a distinct structural change (like the $\beta \rightarrow \alpha$ transition in AgI) but sometimes this is not so clear (as in RbAg_4I_5). Structures which allow fast ion transport are generally disordered, "channelled" or "layered" (Wiedersich and Geller 1971). Ion-Ion interactions or correlation

over, Owens (1971) has noted that M^+ ions with volumes greater than $85(\text{\AA}^3)$ do not generally form conductive compounds. Furthermore, for bigger cations a lesser amount may be needed. For example, the maximum conductivity is obtained with 13 mole % MI for the $(\text{CH}_3)_4\text{NI}-\text{AgI}$ system, 12.5 mole % in the $(\text{CH}_3)_2(\text{C}_2\text{H}_5)_2\text{NI}-\text{AgI}$ system and 12 mole % in the $(\text{C}_2\text{H}_5)_4\text{NI}-\text{AgI}$ system. This means that the percentage of added cations decreases with its increasing size as is expected from Raleigh's picture.

The above structural principles have been deduced on the basis of the very small amount of available structural data — mostly on cation substituted systems. More structural studies would be necessary both on anion and cation substituted systems to arrive at a better understanding.

2.8 Oxygen ion conductors

The first solid oxide electrolyte ever used was probably the ceramic with composition 85 wt % ZrO_2 , 15 wt % Y_2O_3 which was used by Nernst (1899) as an incandescent lighting material. Later Baur and Preis (1937) used this material for a fuel cell. The definite conductivity mechanism in terms of oxygen vacancies was proposed by Wagner (1948) and later verified by Hund (1952). However, the sudden increase of scientific interest in such materials started after Kiukola and Wagner (1957a, 1957b) illustrated their use in high-temperature thermodynamic measurements and fuel cells. In principle, almost all oxides can be expected to show some degree of oxygen ion conduction particularly the non-stoichiometric oxides. The non-stoichiometry can be created by heating the metal oxides in vacuum or oxygen or the relevant metallic vapour (Wagner 1974). Most of the oxygen ion conductors are good only at high temperatures ($\sim 1000^\circ\text{C}$) and are mixed conductors (ionic + electronic + electron-hole). As in the case of other ionic conductors, the conductivity in this case is strongly dependent on the temperature and doping with aliovalent impurities (like Ca^{2+} , Y^{3+} , Sr^{2+} etc. in HfO_2 , CeO_2 etc.) which control the number of point defects and their mobility. However, a unique property of oxide conductors is the dependence of the conductivity on the ambient pressure or activity. If the ambient oxygen pressure is low, the oxygen ions (O_O) would leave the solid electrolyte according to the following mechanism:

The defects are expressed in the Kröger-Vink notation: the symbol indicates the defect; the subscript the location of the defect; and the superscripts (') (.) denote negative and positive charges respectively. For example V_O means vacancy at the oxygen site having effectively two positive charges. Similarly, O_i'' would indicate an oxygen ion at the interstitial site with effectively two negative charges.

tions, cooperative hopping or correlation effects play important roles. Details of the conductivity mechanism will be discussed in Chapter 4. Figure 2.46 gives a schematic plot of conductivity as a function of partial pressure of ambient oxygen for a typical oxide electrolyte at three different temperatures: The higher the temperature, the lower is the range of oxygen pressure over which the conductivity is ionic. The dependence of the range of ionic conduction on concentration of aliovalent dopant (like Ca^{2+} or Y^{3+}) is shown in Figure 2.47. The effect of increasing concentration is to broaden the range of ionic conductivity and shift the whole conductivity curve to lower p_{O_2} values.

Most of the useful oxide electrolytes developed so far are based on ThO_2 , CeO_2 , HfO_2 and zirconia, though some other systems have also been described. For an earlier review see Etsell and Flengas (1970). The common structure which sustains high oxygen ion conduction is the "fluorite structure". The fluorite structure for MO_2 (M = metal ion Th, Ce etc) is shown in Figure 2.48. In this structure there are a large number of octahedral interstitial voids. Each metal cation is surrounded by eight

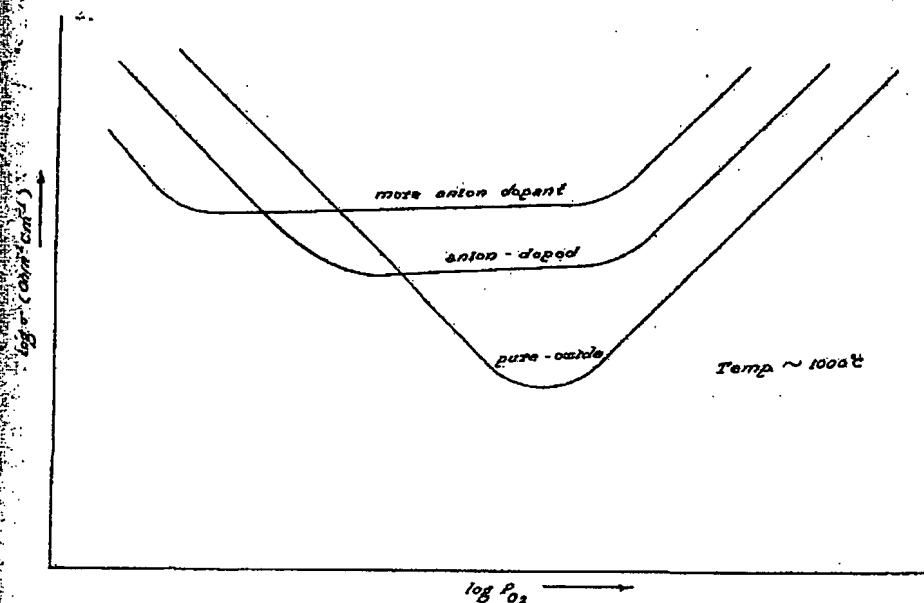


Fig. 2.47 Schematic representation of the dependence of conductivity of any oxide ion electrolyte on oxygen partial pressure for samples doped with different concentrations of aliovalent anion (Ca^{2+} , Y^{3+} etc.).

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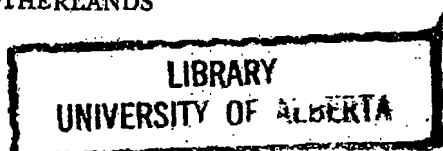
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**MATERIALS SCIENCE
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ENGINEERING
HANDBOOK**

THIRD EDITION

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and
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Electrical Properties**Table 29I. RESISTIVITY OF CERAMICS**

(SHEET 4 OF 6)

Class	Ceramic	Resistivity (Ω -cm)	Temperature Range of Validity
I	Aluminum Oxide (Al_2O_3)	$>10 \times 10^{14}$	25°C
		2×10^{13}	100°C
		1×10^{13}	300°C
		6.3×10^{10}	500°C
		5.0×10^8	700°C
		2×10^6	1000°C
	Beryllium Oxide (BeO)	$>10^{17}$	25°C
		$>10^{15}$	300°C
		$1-5 \times 10^{15}$	500°C
		$1.5-2 \times 10^{15}$	700°C
		$4-7 \times 10^{15}$	1000°C
	Magnesium Oxide (MgO)	1.3×10^{15}	27°C
		$0.2-1 \times 10^8$	1000°C
		4×10^2	1727°C
	Silicon Dioxide (SiO_2)	10^{18}	room temp.
	Zirconium Oxide (ZrO_2)		
	(stabilized)	2300	700°C
	(stabilized)	77	1200°C
	(stabilized)	9.4	1300°C
	(stabilized)	1.6	1700°C
	(stabilized)	0.59	2000°C
	(stabilized)	0.37	2200°C

Compiled by J.S. Park from No. 1 Materials Index, Peter T.B. Shaffer, Plenum Press, (1964); Smithells Metals Reference Book, Eric A. Brandes, ed., in association with Research Institute Ltd. 6th ed. London, Butterworths, Boston, (1983); and Ceramic Society of America (1986-1991).

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Materials for fuel-cell technologies

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Fuel cells convert chemical energy directly into electrical energy with high efficiency and low emission of pollutants. However, before fuel-cell technology can gain a significant share of the electrical power market, important issues have to be addressed. These issues include optimal choice of fuel, and the development of alternative materials in the fuel-cell stack. Present fuel-cell prototypes often use materials selected more than 25 years ago. Commercialization aspects, including cost and durability, have revealed inadequacies in some of these materials. Here we summarize recent progress in the search and development of innovative alternative materials.

The successful conversion of chemical energy into electrical energy to a primitive fuel cell was first demonstrated¹ over 150 years ago. However, in spite of the attractive system efficiencies and environmental benefits associated with fuel-cell technology, it has proved difficult to develop the early scientific experiments into commercially viable industrial products. These problems have often been associated with the lack of appropriate materials or manufacturing routes that would enable the cost of electricity per kWh to compete with the existing technology, as outlined in a recent survey.²

The types of fuel cells under active development are summarized in Fig. 1. The alkaline fuel cell (AFC), polymer-electrolyte-membrane fuel cell (PEMFC) and phosphoric-acid fuel cell (PAFC) stacks essentially require readily pure hydrogen to be supplied to the anode. Accordingly, the use of hydrogen or alcohol fuels requires an external fuel processor to be incorporated into the system. This not only increases the overall efficiency but also increases the complexity of the system. In contrast, solid oxide fuel cells (SOFCs) and molten carbonate fuel cells (MCFCs) operate at higher temperatures, so that they can use hydrocarbon fuels. However, the fuel-processing reaction can be accomplished within the stack, which enables innovative thermal integration/management design features to provide cost-effective efficiencies (>50%).

Although the introduction of a 'hydrogen economy' might seem an attractive scenario, its implementation is beset with formidable technical and economic difficulties. The cheapest technology for the large-scale production of hydrogen is the steam reforming of natural gas, which produces significant emissions of greenhouse gases.³ The type of hydrogen storage is addressed in the accompanying review by Schabert⁴ and Zlot⁵ (see pages 353–366); indeed there is a breakthrough in the production of hydrogen and the development of new hydrogen-storage materials, the concept of a 'hydrogen economy' will remain a utopian exercise. In this article, therefore, we assume that fuel cells have to be designed for operation on hydrocarbon or alcohol fuels to ensure that the technology is able to penetrate the relevant major markets. Otherwise fuel-cell technology will be confined to restricted niche activities where hydrogen might be a convenient option, such as city bus fleets. Clearly the choice of fuel is a

further complication in the factors influencing the commercialization of fuel cells.

Constraints on material selection

Material selection for a commercial product involves an iterative design process that eventually focuses specifically on the particular product and application. However, it is possible to make a few general statements about the selection of materials for fuel cells. The required area-specific reactivity (ASR) of the cell components (electrolyte, anode and cathode) should be below 0.5 A cm² (area density approach), 0.1 (A cm²) (weight approach) and 100 W cm⁻² (area density approach). High power densities are also important to reduce costs, as the amount of material per kW is then minimized. These topics, and considerations of cell efficiencies, are summarized in Box 1.

The need to minimize cell resistance has a major impact on the selection and processing of the cell components. Considerable literature of porous electrode structures was achieved for the first time only about 40 years ago. The electrolyte, gaseous reactants, electrocatalyst and current collectors have to be brought into close contact within a porous structure. The porous structure is typically formed by a combination of hydrophobic polytetrafluoroethylene (PTFE or Teflon) and hydrophilic polyethylene glycol dimethyl ether (PEG or Teflon) greatly simplified the fabrication of porous, high-reactivity porous carbon papers. Most of carbon papers for porous carbon papers provided the electronic pathways, and to facilitate the ASR of the electrode a reliable and reproducible method was usually incorporated into the structure. Further improvements in performance were obtained during the 1970s by depositing small crystallites (2–5 nm) of the electrocatalyst (usually platinum or Pt alloys) onto carbon powder or paper. In retrospect, the successful carbon was probably the first manifestation of an engineered nanostructure, and it is not surprising that its implementation more than 40 years ago was so difficult.

High kinetic conductivities (>1 S cm⁻¹) associated with the liquid KOH, phosphoric acid and molten carbonate electrolytes ensured that, with appropriate design strategies, the ASR values of these components can be small. Although solid-state electrolytes have lower ionic conductivity values, the molten membranes used in the PEMFC system can be fabricated relatively easily as a thin film (100 nm) to produce satisfactory ASR values provided the water content of the film is controlled under the dynamic conditions of cell operation. In contrast, it has been, and continues to be,

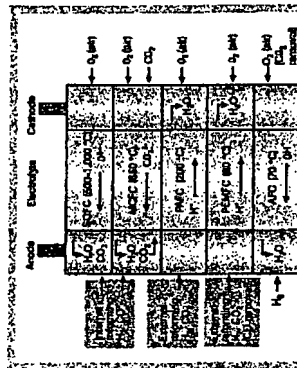


Figure 1. Fuel cell system schematic diagram. The fuel cell system consists of a fuel cell, a power conditioning system (PCS), and a load. The fuel cell has an anode and a cathode. The anode is connected to the PCS, which is connected to the load. The cathode is connected to the PCS, which is connected to the load. The PCS has DC and AC outputs. The load has DC and AC inputs. The fuel cell system is shown in a schematic diagram.

difficult to scale-up thick-film technology to provide cost-effective ceramic multi-cell assemblies comparable with required thicknesses in the range 10–30 μm . Usually, the thick-film electrolyte has to be sintered at temperatures approaching 1,400 °C; this requires a porous ceramic substrate, which is often the anode/cathode material. The substrate material had to be carefully selected to avoid reaction with the electrolyte, and/or thermal expansion mismatch, during the high-temperature sintering process. The incorporation of a relatively weak, brittle structural component in SOFC stacks is at present restricting the application of SOFC systems to those situations that do not demand rapid temperature fluctuations. In this respect the recent development¹ of sintering procedures below 1,000 °C, which should allow the use of metal substrates, represents a significant advance that will enable the development of more rugged SOFC systems.

Another important component in a fuel-cell stack is the impermeable electronic conducting bipolar plate. This has the dual function of distributing the fuel and air to the anode and cathode, respectively, as well as providing the electrical contact between adjacent cells. The corrosive acidic conditions prevailing in the PEMFC and PAFC systems severely restrict the choice of bipolar plate material and at present graphite is usually selected. However, alternative materials or manufacturing methods are mandatory if these systems are ever to sustain the target cost. Major research and development (R&D) programmes are examining the behaviour of alternative carbon-based materials produced by injection moulding, or coated stainless steels. For the high-temperature systems (MCFC and SOFC) operating in the temperature range 500–750 °C, appropriate stainless steel compositions can be specified which seem to satisfy the technical and economic constraints. But for SOFCs operating at higher temperatures (800–1,000 °C), alternative, more expensive bipolar plate materials have to be specified, which at present incur significant cost penalties.

Additional constraints influencing material selection arise from reliability and durability issues. For transport applications, minimal values of performance degradation (for example, 0.1% over 1,000 h) are required for projected operational lifetimes of 5,000 h. But for stationary applications—for example, distributed CHP (combined heat and power) systems—a similar degradation rate must extend over a period of at least 40,000 h (5 years). These different lifetime targets seem to be introducing problems for PEMFC prototype CHP systems, as the stack components were developed originally for transport applications.

A fuel-cell system also incorporates relevant balance-of-plant items such as pumps, valves, heat exchangers and piping. Although these important components contribute at least half the cost of a fuel-cell system, we will not consider them further here, except to note that for the PAFC system, many of which have now been operated for periods approaching 30,000 h, the main source of system breakdown has been balance-of-plant items. External fuel processors (reformers) are also the subject of intensive development around the world, and a variety of innovative compact reformers using diffusion-located pre-reformers, components or micro-channel designs² also illustrate the impact of materials technology on this aspect of fuel-cell systems.

For more than four decades now, reliable, efficient, fuel-cell systems incorporating AFC stacks have proved their worth in the Apollo spacecraft and space shuttles. Excellent electrode kinetics, when operating on pure hydrogen and oxygen, are an attractive feature of this system. But for terrestrial applications, the additional economic penalties, which include the need to replace hydrogen by cheaper hydrocarbon or alcohol fuels, have provided severe problems for materials selection and the associated fuel-processing technology. After 30 years development, the Ebasco consortium abandoned attempts in 1988 to develop a fuel-powered fuel cell for the AEC system. Although Ebasco have purchased the technical rights, it is anticipated that their present generation of AFC systems will be small, in spite of a recent publication³ advocating the use of AFC systems with reformers in the form of hybrid power units.

Approximately 200 PAFC cogeneration units (International Fuel Cells (IFC) FC25, rated at 200 kW) have been installed around the world, and have exhibited excellent reliability. However, the commercial future of this system is possibly in jeopardy as the manufacturers (IFC and Japanese companies) have been unable to reduce the capital cost sufficiently below US\$3,000 per kW, as originally forecast⁴. Most observers⁵ believe that for initial market entry the target cost per kW must be reduced to around US\$1,000 per kW, falling to below US\$500 per kW with volume production. Accordingly, we focus here on materials aspects of the PEMFC, MCFC and SOFC systems, which at present still appear to present opportunities to exploit their potential.

It is important to note that the materials currently being used in PEMFC, MCFC and tubular SOFC prototype demonstration units essentially remain the same as those selected at least 25 years ago. Although innovative fabrication and processing routes have improved the attributes (for example, lower cost and lower Pt loadings) of these materials, it is only in the past five years that system engineering and commercialization teams have highlighted the inadequacies of some of the materials originally selected. As indicated in the next two sections, it is these issues that are now driving the development of alternative materials, particularly for the PEMFC and intermediate temperature (IT)-SOFC stacks.

Polymer-electrolyte-membrane fuel cells
The most important materials under development for PEMFC stacks are construction materials for the cell frames, bipolar plates, electrocatalysts for the fuel and air electrodes, and the ion conducting membrane.

Depending on the fuel to be used in the PEM cell, the requirements for these materials are completely different. The simplest case is the operation with pure hydrogen and oxygen or air. Cells with high



For fuels containing traces of CO, or methanol in the DMFC, a CO-tolerant catalyst is required. This remains one of the most challenging tasks for the successful development of commercial PDMFC

Although the US General Electric Company (GE) initiated the

The following properties of polymeric membranes need to be optimized for use in fuel cells: (1) high proton conduction, assured by acid ionic groups (usually SO_3H), depending on isophonation

There is significant interaction between the desired properties of the membrane — high conductivity, low swelling, low gas and

Because of their PTFE-like backbone and relatively low crystallinity, Nylon and related materials are a favored option in high-temperature applications. They are commonly used in fuel-cell stacks, but the esters remain high on the list of concerns. Therefore, much effort is being applied to the development of cheap, low-cost, usually fluorine-free, membrane materials. But hydrocarbon polymers often suffer from an insufficient thermal stability and to ensure and measure dynamic properties have been introduced into the polymer backbone. Aromatic polymers seem to be the most stable molecules among the hydrocarbons. For example, poly(benzophenone ether sulfone)-based polymers were examined for their use in membranes for fuel cells.

For operation at elevated temperatures, which is desirable for highly power density DMFC systems and for reformers feeds with CO contents above 100 ppm, the conduction mechanism becomes the predominant base in the type of membranes described earlier (hydrocarbonated polymers), the proton-conduction mechanism is based on the migration of hydrated protons. Above 100°C, pressurized operation is required to ensure the presence of liquid water. Phosphoric acid and neutral polymers with unfunctionalized backbones exhibit a conduction mechanism relying on structure dilution¹⁴ and can be used at temperatures above the boiling point of water. As phosphoric acid

It should be emphasized that if an alternative membrane material does emerge, considerable R&D will still be necessary to optimize and manufacture the new membrane-electrode assembly (MEA). This development has taken many years for Nafion-type MEAs, although some of the expertise gained may be able to be transferred to the new system.

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Solid-state fuel cells

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insight review articles

If the SOFC stack is to be integrated with a gas turbine, then system requirements indicate that the temperature of the exhaust gas from the stack should exceed about 850 °C. The steam reformer of such a stack such as diesel, petrol and propane to produce H_2 and CO gases for the waste also, at present, require that processors operating at excess of 700 °C. But it is the properties of the solid electrolyte that exert the biggest influence on stack design and make this selection, as indicated earlier.

Figure 1 summarizes how the specific ionic conductivity of a selection of solid electrolytes varies with the temperature of operation. By taking a typical value of 0.15 S cm⁻¹ for the maximum ASR value, it is possible to calculate the maximum thickness allowed for a given electrolyte component. For example, a design configuration that specified a self-supported (~150 µm) yttria-stabilized zirconia (YSZ) electrolyte would require a temperature of operation greater than 950 °C. Operation at 950 °C poses major problems for planar (flat) SOFC configurations compared to tubular configurations. These include stability of the electrode-electrolyte interface, selection of the bipolar-plate material, and the optimal material composition and arrangement for the seals that are necessary in planar SOFC stacks. Conventional designs are difficult to modify.

Owing to these intrinsic problems with the high-temperature design configuration, there was early interest in tubular designs that eliminated the high-temperature sealing problems. A variety of tubular configurations were initially examined¹, but the arrangement most commonly used by Siemens-Westinghouse has been the most successful. This arrangement uses a 1.5-m porous tubular cathode (La_{0.8}Y_{0.2}Co_{0.8}Fe_{0.2}O₃ or LSCF), after deposition of the La_{0.8}Y_{0.2}Co_{0.8}Fe_{0.2}O₃ (LSCF) cathode, by flame spraying, an electrocatalytic vapor-deposition (VVD) process is used to deposit the porous, thick-layer (30-40 µm) Ni-YSZ anode. The cell structure is completed by using electrolyte (YSZ) bipolar plates. The porous Ni-YSZ waste, although successful in behavior, the VVD process is relatively expensive and efforts are underway to replace this process by an alternative, cheaper fabrication process. However, conventional ceramic routes involving the degradation of YSZ powders and subsequent sintering are constrained by the need to restrict the sintering temperature to below about 1,250 °C to ensure

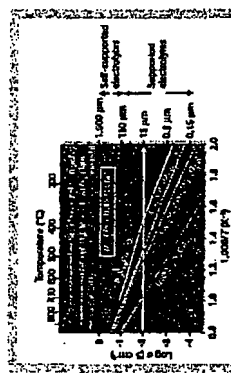


Figure 1. Specific ionic conductivity and thermal expansion of selected electrolyte materials. The graph shows the specific ionic conductivity (Log σ) versus temperature (°C) for various electrolyte materials. The materials shown are YSZ (yttria-stabilized zirconia), LSCF (La_{0.8}Y_{0.2}Co_{0.8}Fe_{0.2}O₃), LSGF (La_{0.8}Y_{0.2}Co_{0.8}Fe_{0.2}O₃), and others. The graph also indicates the temperature required for a self-supported electrolyte thickness of 150 µm.

highly efficient reaction between the LSM cathode and electrolyte. Selection of the cathode (LSM) and anode (Ni-YSZ) compositions was established during the 1970s by Westinghouse and ABB, after examining a variety of oxide composites for long-term compatibility with YSZ at elevated temperatures. This aspect of the work was later explained in terms of an electrochemical model².

The Siemens-Westinghouse tubular design remains the most developed SOFC system, and has been evaluated in turbo-generating systems with microturbines to provide a system capable of generating electricity around 80% efficiency. Large, multi-megawatt integrated systems are predicted to produce electricity at efficiencies approaching 70%. However, these advances are also concerned with the basis of system design rather than the selection and development of new materials.

Microturbine temperature solid-oxide fuel cells. The strategic program of large multilateral companies (such as Westinghouse, GE and ABB) featured the development of multistage, high-temperature SOFC stacks, and these priorities had a strong influence on the development of SOFC design and materials for two decades from 1970 to 1990. However, it was beginning to be recognized that for smaller SOFC stacks not destined to be integrated with gas turbines, the operating temperature should be lowered, and as possible without compromising the electrochemical and thermal performance of the cell. The development of these smaller IT-SOFC stacks for distributed (embedded) CHP units, to provide standby power, is also being stimulated by liberalization (deregulation) of electrical supply policies. In addition, many sustainable microturbines are examining whether small SOFC stacks (3-5 kW) can be developed to supply the electrical power for auxiliary functions such as air conditioning in vehicles.

Examples of the most appropriate solid-electrolyte composition for operation at intermediate temperatures (800-950 °C) can be taken from Fig. 1. It becomes again apparent that the design for compact stacks (for a thick-film thickness of 10-15 µm) the associated operating temperature must be reduced to below 900 °C. For example, the basic conductivity of YSZ at the design temperature of 850 °C is only 0.15 S cm⁻¹ (σ = 1.5 × 10⁻² S cm⁻¹). The design of Fig. 1 indicates that for Co_{0.8}Fe_{0.2}O₃ (LSCF) the relevant operating temperature is about 800 °C, and for La_{0.8}Y_{0.2}Co_{0.8}Fe_{0.2}O₃ (LSGF) the relevant operating temperature is about 850 °C. But it is apparent that the reduction in thickness for dense, impermeable films that can be reliably mass produced using plasma-spray ceramic fabrication routes is around 10-15 µm. The need for a thick-film electrolyte requires the component to be supported on an appropriate substrate. As the substrate is the principal structural component in these cells, it is necessary to optimize the conflicting requirements of mechanical strength and gas impermeability.

Any IT-SOFC configuration that seeks to retain the specific advantages of both the tubular and planar arrangements is being developed by both layers³. This integrated planar-stack concept has provided multi-cell assemblies connected in series and temperature by a ceramic substrate, and has many further features to the original Westinghouse tubular design⁴.

Most development work on planar IT-SOFC systems has involved thick-film YSZ electrolytes, and so far most groups have used anode (Ni-YSZ) substrates, which allow the electrolyte powder to be distributed to a dense film around 1,400 °C. One of the problems associated with using porous, composite Ni-YSZ substrates is their relatively poor thermal-expansion compatibility with the YSZ thick film. Accordingly, several groups are examining porous substrate materials on Ni-YSZ or Ni-TiO₂ compositions, with thin interfacial oxide regions incorporating Ni, YSZ, and/or doped CaO. Although replacement of the YSZ can provide better thermal-expansion compatibility, problems still remain over the volume changes associated

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Abstract. A systematic review of the 1996-1999 meta-CDs found 16 randomised controlled trials (RCTs) comparing the use of a single or double-blind placebo-controlled RCT. The results of the meta-CDs were compared with the results of the RCTs. The meta-CDs were found to be more likely to report a significant effect than the RCTs. The meta-CDs were found to be more likely to report a significant effect than the RCTs. The meta-CDs were found to be more likely to report a significant effect than the RCTs.

weight, the reduction and oxidation of the N component. As the various substituted-ethylene flours are usually co-fired in the municipal incinerator, the presence of NO_x which can be carefully reduced by the use of a selective catalyst, is not a problem. The reduction of the NO_x during the flue's handling cycle of the assimilated stack. Additionally, operating parameters have to be designed to prevent the NO_x from oxidizing when SO₂ is added down to the streamer to prevent the formation of sulfuric acid. The use of a selective catalyst, N₂H₄, to prevent the NO_x from oxidizing to form nitric acid, is not a problem. The use of a selective catalyst, N₂H₄, to prevent the NO_x from oxidizing to form nitric acid, is not a problem.

[illegible][illegible]

Ti-SiO_2 stacks incorporating alternative acceptor/donor. Although YSZ is still the measured electrolyte material for SOFC stacks, solid non-oxide electrolyte material is not without problems and research

continues into the long-term evaluation of scandia-doped ZrO_2 , which can exhibit higher ionic conductivities than the traditional YSZ material.

In principle, the use of cast-based electrolytes is limited to low operating temperatures to be applied to around 500 °C (see Fig. 4). But porous-based electrolytes associated with PEN (see Fig. 4) that porous-based electrolytes have restricted the investment in this technology. It is well known that, at elevated temperatures, O^{2-} ions can be reduced to $\text{O}^{\cdot-}$ under the rich-oxygen conditions prevailing in the anode compartment. The associated electrochemical conductivity (and deleterious by the dependent) produces an internal short circuit in the PEN structure, which can significantly degrade the efficiency and performance of the device. However, the operating temperature is lowered to around 300 °C, then the electrode material stability is not so dependent under this operating temperature.

Another significant difficulty that has restricted exploitation of the attractive properties of NiO at 500°C has been the need to develop an alternative cathode composition that functions effectively at these temperatures. Recent developments in this area have been reported by Kojin et al.¹⁴ and there are indications that this approach to maintain air cathodes can be functional which exhibit small overpotentials at 500°C (for example, 0.15 V at 1 A cm⁻²). Cathodic materials such as Ni-20Co also provide adequate performance at 500°C for humidified oxygen fuels, indicating that, for example, Ni-20Co¹⁵ stacks at 500°C are now a viable option.

[illegible]

Operation at 500 °C allows the use of compliant high-temperature guides in place of rigid, brittle glass or ceramic rods, thus permitting greater design flexibility for the reactor configuration. At Imperial College, London, researchers have taken advantage of the fact that the thermal-expansion coefficient of CGO and ferritic stainless steel are virtually identical ($12.5 \times 10^{-6} \text{ K}^{-1}$), so that the thick-film IPN structure can be structured on robust stainless steel. These metal-supported IPN structures are robust, and should withstand the rapid temperature cycles expected during operation of full-size SOFC stacks.

Another electrolyte, liquid LiClO_4 (ESQM), is also attracting much attention for Li-ion battery applications. Although its conductivity is slightly smaller than that of LiPF_6 , it is more stable. Its decomposition is slightly smaller than that of LiPF_6 at 500°C. Its acid content is a weaker acid than that of LiPF_6 and it is more appropriate to use this electrolyte at temperatures around 0-60°C, where the reduction of Li^+ and CO_2 takes place more significantly. It has been difficult to fabricate pure high-phases ceramic electrolytes, and second phases such as Li_2CO_3 and Li_2O in LiClO_4 are often detected in the grain boundaries. Whether these phases are responsible for the enhanced reactivity of ESQM, or whether it is an intrinsic property of ESQM, are questions that require urgent answers. Moreover, the preferred composition, $\text{Li}_x\text{M}_y\text{O}_z$ ($\text{M} = \text{Co}, \text{Mn}, \text{Ni}$), does not seem to be stable at lower temperatures (P. M. Shewchuk, personal communication). Although research cautions into the synthesis of alternative organo-bonded electrolytes, it has proved difficult to prepare alternative materials with an appropriate balance of properties that can displace the traditional carbonate-based electrolyte LiPF_6 in EC/DEC .

[illegible]

Spar Plug use alumina since it is electrically insulator:

APPENDIX E

<http://yshsp.en.alibaba.com/search/offer>

Auto Ignition System use alumina since it is an insulator:

<http://starsparkplug.en.alibaba.com/offerlist.html>

Alumina insulating coating:

<http://www.freepatentsonline.com/6844023.html>

Shanghai Morgan Matroc Technical Ceramics Co.,Ltd

High efficiency alumina heat-electric Insulator for welding and heat treatment. Morgan Technical Ceramics Shanghai produces a complete range of ceramic ...
www.morganmatrocsh.com/e_chanpin_7.htm - 15k - [Cached](#) - [Similar pages](#)

Ortech industry -table containing alumina 's properties:

<http://www.ortechceramics.com/alumina.htm>

Ceramic Tube and Ceramic Rod Products - (111 companies)

Ceramic tube and ceramic rod products are suitable for use in applications requiring high temperature strength, erosion resistance, electrical or thermal insulation, and other specialized characteristics. [Search by Specification](#) | [Learn more about Ceramic Tube and Ceramic Rod Products](#)

Alumina Insulator:

<http://www.prgtech.com/engineering.html>

Alumina use in thermocouple assembly as a insulator:

PDF Noble Metal

File Format: PDF/Adobe Acrobat
elements, elements with insulators or assemblies. A typical assembly includes a head, alumina insulators and a protecting ...
www.watlow.com/literature/specsheets/files/sensors/ricnom0305.pdf - [Similar pages](#)

Page 8 -shows use of alumina tube in an assembly as a insulator:

Henry Rohrs poster

File Format: PDF/Adobe Acrobat
Alumina Insulator. Nickel Seat. Gold O-Ring. SS O-Ring Seat. Vespel Support. Sapphire Ball. PZT Bimorph. Pulsed Valve. The pulsed valve consists of a ...
www.chemistry.wustl.edu/~msf/ASMS98/Pshenry98.pdf - [Similar pages](#)

Goodfellow- a table listing volume resistivity of alumina $>10^{14}$ ohm-cm.:

<http://www.azom.com/details.asp?ArticleID=2103>

APPENDIX F

There are no related appeals, interferences, or judicial proceedings.